The Effects of Tungsten's Pre-Irradiation Surface Condition on Helium-Irradiated Morphology

Lauren M. Garrison^{a,b*} and Gerald L. Kulcinski^a

^aUniversity of Wisconsin, 1500 Engineering Drive, Madison, WI 53706-1609, USA

^bPresent address: Oak Ridge National Laboratory, One Bethel Valley Road, Oak Ridge, TN 37831-6133, USA

*Corresponding author: garrisonlm@ornl.gov

Abstract

Erosion is a serious concern associated with the use of tungsten as a plasma-facing component in fusion reactors. To compare the damage progression, polycrystalline tungsten (PCW) and (110) single crystal tungsten (SCW) samples were prepared with (1) a mechanical polish (MP) with roughness values in the range of 0.018–0.020 μm and (2) an MP and electropolish (MPEP) resulting in roughness values of 0.010–0.020 μm for PCW and 0.003–0.005 μm for SCW samples. Samples were irradiated with 30 keV He^+ at 1173 K to fluences between 3×10^{21} and 6×10^{22} He/m^2 . The morphologies that developed after low-fluence bombardment were different for each type of sample—SCW MP, SCW MPEP, PCW MP, and PCW MPEP. At the highest fluence, the SCW MPEP sample lost significantly more mass and developed a different morphology than the SCW MP sample. The PCW samples developed a similar morphology and had similar mass loss at the highest fluence. Surface preparation can have a significant effect on post-irradiation morphology that should be considered for the design of future fusion reactors such as ITER and DEMO.

Keywords: plasma-material interaction; plasma-facing material; fusion materials; sample polishing; tungsten; helium ion irradiation

1. Introduction

Although tungsten has been selected as the divertor material for ITER [1, 2], was the choice for the first wall of the High Average Power Laser (HAPL) reactor design project [3], and will likely be a plasma-facing material for future fusion DEMO reactors, there are several variables to be decided for producing tungsten components. Different methods of manufacturing, different crystallographic textures, and different tungsten geometries can each lead to varied responses of tungsten to fusion conditions. In addition, the surface preparation of tungsten can play an important role, but it is often not the focus of experiments simulating fusion conditions. Reviewing recent papers on plasma-material interactions reveals a variety of different sample preparation techniques have been used. Ueda et al. prepared tungsten samples by mechanically polishing them to a mirror finish with 10 nm roughness before annealing them at 1173 K for 30 minutes for stress relief [4]. Baldwin and Doerner mechanically polished tungsten samples to below 50 nm roughness but did not heat-treat them [5]. Ryazanov et al. specifies only that the tungsten samples were “polished and cleaned” [6]. Zenobia et al. used a mechanical polish to unspecified roughness followed by electropolishing with 2 wt % KOH and a 15 minute anneal at 1173 K [7]. Each of these treatments alters the material's near-surface microstructure in different

ways, which may in turn result in different irradiated microstructures, thus making it difficult to compare results from the separate experiments.

In certain materials, such as silicon, polishing can create an amorphous layer on the surface [8]. An amorphous layer is never formed on mechanically polished metals, but mechanical polishing can cause drastic changes to the near-surface region as compared to the bulk [9]. The subsurface damage layers from polishing [10] are essentially the same as those created in a sliding or wear test [11] except in terms of scale.

A key reference in understanding the changes to the near surface region because of polishing is the book *Metallographic Polishing by Mechanical Methods* by Samuels [12], and much of the following discussion is based on his work. A standard first polish for tungsten is accomplished by using a polishing pad with embedded abrasive particles such as silicon carbide. If the pressure is high enough, when the tip of an abrasive particle contacts the sample surface, it creates plastic strain in a region larger than the size of the tip [13]. As the polishing pad slides past the sample, the abrasive tip can either remove a miniature machining chip from the sample surface or dig a furrow, depending on the tip's angle to the sample surface [14]. The sample first accommodates the strain from the abrasive particle by slipping along a slip system, and as the strain increases, a slip band forms to start the chip removal from the surface [10]. The slip motion creates sub-grains surrounded by dislocations in the near surface region [10] in orientations that allow further shear motion [13]. For tungsten, the primary slip systems are $\{110\}\langle 111 \rangle$ and $\{112\}\langle 111 \rangle$ [15].

Typically, this stage of polishing is done with a machine that rotates the sample, the polishing pad, or both, so the abrasive particles scrape the sample surface in many different directions. This prevents the near surface region from uniformly rotating to a single, preferred slip system as might occur in a sliding test. As a result of the polish with abrasive pads and the rotation, the sample develops a highly damaged and strained layer of small grains, called the shear band layer, at the surface [10]. Below that layer is the deformed layer which may have altered grain structure from the bulk material and also has strain that decreases with depth until the elastic-plastic boundary, below which the bulk material is unaffected by the polishing [10].

After using abrasive polishing pads, a sample will be further polished with loose abrasives, such as diamond powder, in liquid or paste. Diamond powders damage the sample surface by essentially the same mechanism described above for abrasive pads but on a smaller scale [9]. However, at diamond sizes of 0.1 μm and below, as well as for colloidal silica polishing, the mechanism of polishing changes and is not fully understood [16].

Samples that are mechanically polished always have scratches, although the size reduces with finer grit of polish [16], and they always have strain in a subsurface layer [9] because of the mechanism of material removal from the surface. The depth of the damaged and strained layer that remains on the sample after polishing depends on the sample characteristics, abrasive characteristics, pressure used in polishing, and whether or not enough material was removed by each polishing step to physically remove the damage layer left by the previous polishing step. For example, a 30% Zn brass polished with 220-grade silicon carbide paper had a shear band layer on the order of 3 μm and a deformed layer on the order of 40 μm [10] while the same type of material polished with 1 μm diamond paste developed a shear band layer approximately 0.1 μm and deformed layer of 0.7 μm [9].

In contrast, removing surface material by chemical methods, such as electropolishing, does not create any strain in the material and can remove the surface layer damaged by a mechanical polish, resulting in a surface microstructure that is more representative of the bulk

material [17]. Artifacts can be created by electropolishing, for example if the chemical preferentially attacks grain boundaries, but it does not create any plastic strain in the sample [9]. Additionally, while the material removal rate of refractory metals from abrasives is slow [18], chemical reactions with aqueous NaOH or KOH easily dissolve tungsten metal [15].

Because these different preparation methods result in different microstructures on the surface of the material, they may result in different microstructures after irradiation. Different surface finish techniques should be selected based on the aim of the study—to simulate what will happen in reactor, it may be beneficial to use the same mechanical polish that will be used on a real component. However, to investigate the science of gas interaction with tungsten surface, a well characterized surface without strain that is representative of the bulk microstructure, as created by electropolishing, may be best. If different surface finishes result in different irradiated morphologies, different amounts of mass loss, or different gas trapping characteristics then the surface finish should be optimized for use in fusion reactors.

To address questions of whether surface preparation differences can cause irradiated morphology differences, two groups of samples were prepared. All samples were mechanically polished (MP), and then half of the samples were additionally electropolished (EP). Samples with different surface finishes were irradiated to identical conditions in the Materials Irradiation Experiment (MITE-E) at the University of Wisconsin Inertial Electrostatic Confinement (UW-IEC) Laboratory. By removing surface preparation as a variable, this experiment also allows closer comparison between samples irradiated in the MITE-E and those previously irradiated in the HELIOS UW-IEC device.

2. Methods

Samples of polycrystalline tungsten (PCW) and single crystal tungsten (SCW) with surface normal orientation (110) were obtained from Oak Ridge National Laboratory, where they had been cut to size: $\sim 10 \times 10 \times 1$ mm for the PCW and ~ 8 mm diameter $\times$ 1 mm thickness for the SCW. Samples were MP using silicon carbide polishing pads in grades 800, 1200, and 2400. Then, samples were polished with diamond suspensions of 6, 3, and 1 μm . The final step in the MP procedure was a 2% ammonium hydroxide, 2% hydrogen peroxide, and 96% colloidal silica solution. Although the colloidal silica solution is estimated to polish a surface with a combination of chemical and mechanical means [16], the resulting finish on the samples appears to be dominated by the abrasive steps, so this whole procedure is labeled the “mechanical polish”. Selected samples of PCW and SCW were then EP with 2 wt % KOH and de-ionized water at the UW-IEC Laboratory.

A SCW sample that was MPEP, a PCW sample that was MPEP, and a PCW sample that was MP were scanned with a Zygo NewView Interferometer to analyze the roughness of the surfaces. Surface roughness is not necessarily related to the depth of the damaged and strained layer from MP and gives no information about if the surface is representative of the bulk crystal structure. However, certain ion bombardment phenomenons such as sputtering are sensitive to the initial roughness of the sample. Multiple spots on each sample were imaged in the interferometer using the highest magnification setting, which examines an area 70 by 50 μm . Fig. 1 shows the interferometer scan of each sample on the spot with the lowest roughness, as well as the range of measured roughness values for each sample. The SCW MPEP sample (Fig. 1a) has a uniform surface finish with slight deviations in height and has the smoothest surface of the three by almost an order of magnitude. The line scan of the SCW sample (bottom image in Fig. 1a)

shows the surface height varies from -0.01 to $+0.01$ μm without substantial dips or peaks. The PCW MPEP sample has a greater overall surface roughness than the SCW that received the same treatment; and the PCW sample's surface finish is less uniform, with isolated deep pits and peaks on the surface (Fig. 1b). The PCW MP sample's roughness values calculated from the scan (Fig. 1c) are lower than those of the PCW MPEP sample, but the roughness is distributed differently on the two samples. The PCW MP sample has hills and valleys uniformly distributed over the whole surface, whereas the PCW MPEP sample has large smooth regions with isolated peaks and valleys. Based on scanning electron microscope (SEM) analysis, the surface states of the PCW MP and SCW MP samples are almost indistinguishable, so it is inferred that the roughness values are also similar. This observation matches that of Rigney and Glaeser who note that because of the plastic deformation and slip, the surface layer of a polished sample becomes highly damaged, as compared to the bulk crystal structure, and once steady state is reached may be identical for materials whose bulk is single crystal or polycrystalline [17].

Fig. 1. Surface roughness images from the Zygo NewView Interferometer for samples before irradiation that received different surface treatments: (a) single crystal tungsten mechanically polished and electropolished (MPEP), (b) polycrystalline tungsten MPEP, and (c) polycrystalline tungsten mechanically polished.

The irradiation conditions of the samples are shown in Table 1. All the samples were irradiated in the MITE-E at 1173 K with 30 keV $^4\text{He}^+$ at a nominal ion flux of $9 \times 10^{18} \text{ He}^+ / (\text{m}^2 \text{s})$. The chosen energy is higher than the energy of most particles that will impact the ITER divertor, typically energies < 400 eV [19], but the divertor will experience some charge exchange neutrals with tens of keV energy. For an inertial fusion reactor such as HAPL, 30 keV falls within the expected energy spectrum for particles incident on the first wall [20]. This temperature and energy were also chosen to allow the best comparison with previous experiments at the UW-IEC Laboratory. The ion beam of the MITE-E is known to have a higher flux at the center of the ~ 8 mm beam, and the profile is estimated to be a Gaussian shape. Except where noted, the fluences listed are the average fluence across the 8 mm damaged zone of the sample, and images are from the center of the damaged region. For all samples, the fluence at the center is estimated to be 4.6 times higher than the average fluence. The MPEP samples are referred to here for comparison

with the MP samples; more detail on the SCW MPEP sample set can be found in [21], on the PCW MPEP sample P29 in [7], and on the PCW MPEP samples P37 and P39 in [22].

Table 1: Irradiation conditions of the samples at 1173 K. MP = mechanical polish; MPEP = mechanical polish and electropolish.

Sample name	Fluence (He/m ²)	Irradiation time (min)	Pre-irradiation treatment
S23	3.1×10^{21}	5.0	MP
S22	1.0×10^{22}	18.2	MP
S25	6.0×10^{22}	106.5	MP
P42	1.0×10^{22}	18.5	MP
P43	6.0×10^{22}	104.0	MP
S20	3.0×10^{21}	5.7	MPEP
S16	1.0×10^{22}	18.0	MPEP
S18	6.0×10^{22}	110.8	MPEP
P29	1.0×10^{22}	19.0	MPEP
P37	3.0×10^{22}	54.5	MPEP
P39	6.0×10^{22}	104.2	MPEP

Samples were imaged with a LEO 1530 field emission SEM at the University of Wisconsin Materials Science Center. Cross sections of samples were ion milled using a Zeiss 1500XB CrossBeam Workstation at the Materials Science Center. All samples were weighed before and after irradiation using a Sartorius CPA26P microbalance. Electron backscatter diffraction (EBSD) scans were performed using the EDAX TSL Orientation Imaging Microscopy version OIMDC data collection 5.31 program. The EBSD data were analyzed using the corresponding EDAX TSL Orientation Imaging Microscopy version 5.31 package 7000.

3. Results and Discussion

3.1. Single Crystal Tungsten (110)

Figure 2a is an example of an MP sample surface before irradiation. Scratches of varying depths across the sample surface remained from the polishing. Based on the scratches left on the surface after MP, it appears the tungsten surface deformed plastically when miniature chips were removed by the abrasive polish rather than forming pitting as is seen in polishing some brittle materials [23]. Figure 2e is characteristic of the state of the single crystal MPEP samples before irradiation. Overall, EP gave a smooth surface finish with scattered protuberances on the order of 100 nm. EBSD analysis performed on samples after MPEP confirmed that the surface orientation was (110). EBSD could not determine the orientation of an MP sample because the top region of the sample was damaged by the MP, but the MP samples were from the same batch of (110) samples as the MPEP samples. The presence of the scratches and inability of the EBSD to determine the crystal orientation of the MP samples indicates that the surface of the MP samples may be composed of sub grains in a shear band layer, as predicted by the mechanical polishing model [10], and is not representative of the single crystal of the bulk sample.

Fig. 2. Comparison of samples that received mechanical polishing (MP) before irradiation—(a), (b), (c), and (d)—with samples that received mechanical polishing and electropolishing (MPEP) before irradiation—(e), (f), (g), and (h). All samples were single crystal tungsten with (110) orientation and were irradiated with 30 keV He⁺ at 1173 K. All scale bars are 100 nm.

The MPEP sample irradiated to the lowest fluence, Fig. 2f, had tiny ripples with wavelengths of ~20 nm and ~30 nm diameter pores covering the surface. EBSD analysis of the irradiated surface of S20 revealed that the ripple ridges were aligned with the $\langle 001 \rangle$ direction (within the 5° tolerance of the EBSD data analysis) on the (110) surface, illustrated as a ball model in Fig. 3. Although ripples are commonly seen on crystalline materials irradiated at an angle to the surface, when the irradiation is at normal incidence, the atom arrangement on the surface plane primarily determines the direction of the ripples [24]. It might be expected that the ripple ridges would form in alignment with the close-packed $\langle -11-1 \rangle$ direction because of the ease of diffusion in that direction. However, at 1173 K, diffusion is not limited to nearest-neighbor jumps; in fact, jumps in the $\langle 001 \rangle$ and $\langle 1\bar{1}0 \rangle$ directions could be the primary motion[25]. This trend of directional structures forming on tungsten (110) surfaces may be independent of the helium energy. Nishijima et al. irradiated an EP (110) SCW sample with helium at only 25 eV at 1650 K, and the surface developed a wavy, porous structure with wavelengths on the order of ~40 nm [26].

In contrast, the MP sample S23 at the same fluence, Fig. 2b, developed large ripples with wavelengths on the order of 100–200 nm and individual scattered pores. Also, the MP sample S23 morphology was not uniform across the surface but was a patchwork of two different types (Fig. 4). The large ripples covered both the higher and lower regions of the surface on sample S23 and had boxy edges rather than being thin grass-like tendrils (Fig. 4).

Fig. 3. A ball atom model of the (110) BCC surface created with the Surface Explorer application [27]. Ripple ridges on (110) SCW sample S20 irradiated to 3.0×10^{21} He/m² with 30 keV He⁺ at 1173 K were parallel to the <001> direction.

Fig. 4. Sample S23, SCW (110), was mechanically polished before being irradiated to 3.1×10^{21} He/m² with 30 keV He⁺ at 1173 K. Large ripples cover a surface that has the appearance of forming grains.

When the fluence was increased to 1.0×10^{22} He/m², the MPEP sample S16 had a sponge-like morphology with pores, on the order of 100 nm in diameter, surrounded by thin tendrils of material (Fig. 2g). At the same fluence, the MP sample S22 (Fig. 2c) again had a patchwork of two different surface morphologies, shown in more detail in Fig. 5 beside the MPEP sample S16. Whereas the morphology of MPEP sample S16 was uniform over the irradiated area, MP sample S22 had two morphologies in different regions: one structure of uniform pores with an average size of 42 nm (Fig. 5d) and a second structure with pores of a similar size but a faceted appearance (Fig. 5c). Because S16 and S22 were irradiated to identical conditions, it appears that pre-irradiation surface treatment had an important influence on the resulting structure.

More insight into the structure of S22 was gained from examining its focused ion beam (FIB) cross section in Fig. 6a. The microstructure distinguished by a uniform distribution of small pores (Fig. 5d) is seen in Fig. 6a to be about 250 nm higher than the other microstructure composed of tilted pointed structures (Fig. 5c). Sample S16 did not have height variations like those seen on sample S22 in Fig. 6a.

Fig. 5. Both samples are single crystal (110) tungsten and were irradiated to $1.0 \times 10^{22} \text{ He}^+/\text{m}^2$ with 30 keV He^+ at 1173 K in the MITE-E. (a) Sample S16 was MPEP before irradiation and developed a uniform morphology over the surface. (b) Sample S22 was MP before irradiation and developed two distinct morphologies that are shown in more detail in (c) and (d).

Fig. 6. Both single crystal tungsten (110) samples were mechanically polished before irradiation to (a) $1.0 \times 10^{22} \text{ He}^+/\text{m}^2$ and (b) $6.0 \times 10^{22} \text{ He}^+/\text{m}^2$. The samples are tilted at 54° and image tilt correction on the SEM allows accurate height measurements perpendicular to the sample surface.

It might be expected that the MP S22 with pre-irradiation roughness values four to five times that of the MPEP samples would form ripples more readily, because other researchers have found that ripples form at lower fluences or with larger amplitudes on rougher surfaces [28, 29]. However, the resulting structure on S22 appears to be more influenced by how the MP changed the grain structure rather than by the roughness of the top layer. The 250 nm tall upper layer seen in Fig. 6a may correspond to the layer damaged by MP, and the lower area may be the revealed underlying (110) crystal.

To identify the crystal orientation of the different morphologies on S22, an EBSD scan was performed on a zone of the sample (Fig. 7). The scan was performed with a $0.2 \mu\text{m}$ step size, and the data underwent a grain dilation cleanup with 5° tolerance angle. Although some points remain unidentified, further attempts to clean up the data obscured the interesting features. The EBSD scan revealed that all of the higher plateau regions with uniform pores (Fig. 5d) were close to a (111) orientation, whereas the lower faceted regions (Fig. 5c) were closer to a (110) orientation. Following a path on the sample starting from the highest area at the center of the sample, point A, and ending at the lowest area of the sample, B, in Fig. 7, it is observed that the general orientation color changes from dark blue to light blue to light green. Matching those colors with the inverse pole figure legend in the corner of Fig. 7, it is as if one were tracing from the (111) corner along the edge to the (101) green corner. This may indicate that MP rotated the

top layers of the sample toward a (111) orientation while the underlying sample orientation remained (110). While MP is known to rotate sections of crystals [30] or break the surface into smaller grains [10], the MP-created sub grains tend to align with a slip system of the material, which would be $\{110\}\langle 111\rangle$ or $\{112\}\langle 111\rangle$ for tungsten [13], not the observed (111) orientation. Also, since (111) orientation was not measured on the MP samples before irradiation, it is postulated that the MP created a disordered layer of rotated sub grains, but that the ion irradiation at high temperature caused the formation of the (111) orientation upper layer.

Since (110) is the most closely packed plane of BCC crystals [31], it is interesting that the lower region was not flat but rather appears to be faceted in another, less closely packed direction. Faceting from (110) to ridges with (111) sides has been observed on BCC iron with antimony on the surface [32] but tungsten single crystals of (110) were resistant to faceting even with oxygen present on the surface [33]. It is also possible that the lower region is not, in fact, the perfect (110) lattice, but remains partially damaged or rotated by the polishing.

Fig. 7. SEM image (top) and EBSD map (bottom) show the same area on Sample S22, single crystal tungsten with (110), which was mechanically polished before irradiation to 1×10^{22} He^+/m^2 with 30 keV He^+ at 1173 K.

At the highest fluence of 6.0×10^{22} He/m^2 , the MPEP sample S18 had deeper, larger pores on the order of 100–150 nm surrounded by smaller 30–50 nm pores (Fig. 2h). The fibrous structure seen on the lower-fluence samples had presumably all been sputtered away, leaving only the deep pores. Looking within the larger pores, some ridge-like structures can be seen. It is possible that the porous layer existed on top of a layer that had formed ripples. Instead of large pores on top of a ripple structure, MP sample S25 developed ripples in a uniform direction over the whole irradiated zone (Fig. 2d). The ripple structure has been referred to as a “grass” structure when observed previously on MPEP PCW samples irradiated at similar fluences and temperatures in the MITE-E [7]. Within the grass structure, small ~10–20 nm pores are also visible.

The SCW MP sample S25 irradiated to the highest fluence did not have the two separate damage regions that were observed on the two lower-fluence MP samples, S23 and S22. Instead, a uniform grass structure existed over the entire irradiated zone. The ripples were oriented in the same general direction everywhere, but they were not perfectly straight. An EBSD scan was unable to determine the orientation of the ripples, likely because of the pointed shape of the features and the height differences of the morphology. Viewed from directly above, the sample features appear as grass, as seen in Fig. 2d. However, when the cross section of the same area is viewed, the morphology appears to consist of clusters of pointed features rather than smooth

rows of grass (Fig. 6b). Gaps can be seen between the individual needles, but within each needle there are smaller helium bubbles. The subsurface semi-porous layer extends ~ 730 nm below the surface on S25. The MP sample S25 was irradiated to a fluence six times higher than was MP sample S22, but the damage on sample S25 was almost three times as deep as that on sample S22 (Fig. 6). The depth of the visible subsurface bubble layer on the MPEP sample S18, irradiated at the highest fluence, is only ~ 90 nm [21]. Although the surface view of sample S18 (Fig. 2h) suggests there could be a ripple structure underneath the porous top layer, evidence of such a structure could not be seen in the FIB cross-section view [21]. The MP sample S25, which was irradiated to the same fluence and temperature as S18, had a damage layer over eight times deeper than the layer on S18. This result indicates that the MP pre-irradiation surface finish may allow helium deeper beneath the surface of the sample than does MPEP pre-irradiation surface treatment. This is the opposite behavior from that predicted by calculations of helium penetration depths. In an SRIM (Stopping Range of Ions in Matter) calculation, which assumes an amorphous target, similar to the disordered shear band layer of the MP surface finish, the peak helium concentration is ~ 60 nm below the surface; whereas in a model of helium penetration into a (110) surface, similar to the MPEP surface state, the peak helium concentration, linearly interpolated for 30 keV He^+ implantation, is on the order of 400 nm below the surface [34]. The experiment and model differences are likely explained by the mass loss of the samples; penetration depth calculations do not account for changes in the surface level of the sample during implantation. In the samples irradiated in this work, it is thought that the original surface was eroded to a lower level because of the measured mass loss, discussed in Section 3.4.

3.2 Polycrystalline Tungsten

Before irradiation, the MP PCW surface state (Fig. 8a) is similar to the MP SCW surface state (Fig. 2a). The difference in the surface condition of the MPEP PCW (Fig. 8e) and SCW samples (Fig. 2e) is the visible presence of grain boundaries on the PCW samples. At the lower fluence of $1.0 \times 10^{22} \text{ He}^+/\text{m}^2$, the MP PCW sample P42 developed patches of different morphologies (Fig. 8b) just as the MP SCW sample S22 irradiated to the same conditions did (Fig. 5b). There is on the order of 350 nm between the higher and lower regions of MP PCW sample P42 (Fig. 9a); both regions are porous, and neither appears faceted as does the lower region on MP SCW sample S22 (Fig. 5c). Before irradiation, the MPEP PCW samples had exposed grains with isolated pits and peaks (Fig. 8e). At the lower fluence of $1.0 \times 10^{22} \text{ He}^+/\text{m}^2$, the MPEP PCW sample P29 grain boundaries are easily visible and the morphology consists of the early stages of the grass and needle morphologies seen previously on samples irradiated to those conditions in the MITE-E [7]. The height differences among grains on P29 were significantly less than the 350 nm observed on P42, and the grains were much more defined.

Fig. 8. Comparison of samples that received only mechanical polishing (MP) before irradiation (a, b, c, and d) with samples that received mechanical polishing and electropolishing (MPEP) before irradiation (e, f, and g). All samples are polycrystalline tungsten and were irradiated with 30 keV He^+ at 1173 K. All scale bars are 1 μm except lower image in (d). Average fluences are listed in the figure. All images are from the center of the irradiation zone except (c), which is an area of P43 ~ 1.6 mm from the center of the irradiated zone that received an estimated 1.3×10^{23} He^+/m^2 local fluence, compared with (d) at the center of P43, which received an estimated 2.7×10^{23} He^+/m^2 local fluence.

Fig. 9. Height of structures observed on PCW samples mechanically polished (MP) before irradiation (a, b, and c) and mechanically polished and electropolished (MPEP) (d). All samples irradiated with 30 keV He^+ at 1173 K. Cross-section cuts were made with the FIB and images viewed in the SEM. Samples were tilted 54° , and the SEM image tilt correction was used to correctly measure heights.

On MP sample P43 irradiated to an average fluence of 6×10^{22} He^+/m^2 , it appears that the upper layer visible on P42 continued to be sputtered away to reveal a grass morphology underneath. At about 1.6 mm from the center of the irradiated zone on P43—which experienced a local fluence of approximately 1.3×10^{23} He^+/m^2 —the remains of the plateaus that are visible on P42 appear as islands of smeared material from the MP, and the lower morphology is revealed to be a grass structure (Fig. 8c).

At the center of the irradiated zone on sample P43 (Fig. 8d), which received an estimated local fluence of $2.7 \times 10^{23} \text{ He}^+/\text{m}^2$, the islands of smeared material were not present. It is assumed that since the center of the irradiated zone experienced a higher fluence, all the islands seen in Fig. 8c were either sputtered away or absorbed into the grass structure. There were remnants of the top layer, but they appear as porous confetti distributed over the top of the grass structure. Interestingly, many of these pieces appear to be needle-shaped; but the tips have broken off from the bases, as seen in the higher magnification image below Fig. 8d. Most of the needles are fragmented, but some are as long as 800 nm. It is unknown whether the ion irradiation or the handling of the sample after irradiation caused the needles to break. Pores are visible on the surface of and within the needles. Both the MP SCW and MP PCW samples at the highest fluence of $6.0 \times 10^{22} \text{ He}^+/\text{m}^2$ developed a similar grass structure at the center of the irradiated zone. On the SCW sample, S25, the grass structure is essentially uniform in direction across the sample surface; whereas on P43, isolated ripples are aligned, but there is no overall order.

The main feature on both PCW MP sample P43 (Fig. 8d) and PCW MPEP sample P39 (Fig. 8e) irradiated to the highest fluence of $6.0 \times 10^{22} \text{ He}^+/\text{m}^2$ at 1173 K is the grass structure, and regions can be found on the two samples that are nearly identical. For the MPEP sample P39, at this high fluence the grain boundaries are less distinct and much of the grass has lost its clear directionality. Still, some regions with parallel ripples are distinguishable. For the MP sample P43, the grass direction was also less ordered. However, the extra feature on P43 that was not present on P39 was the over-layer of broken needles.

An EBSD scan on P43 identified each data point, $0.2 \text{ } \mu\text{m}$ step size, as a different orientation; but the grain size of the bulk material of the PCW samples is known to be on the order of a few micrometers. It is possible that shear band layer from the MP before irradiation was composed of very small sub-grains of different orientations, and these remain after irradiation.

The height of morphology varied from grain to grain on both MP sample P43 and MPEP sample P39. The tallest features on P43 of the regions surveyed were needle structures on the order of 830 nm tall (Fig. 9b), while other areas had features 480 nm tall (Fig. 9c) and other areas had shallower features. When viewed at higher magnification, individual helium bubbles can be seen in the cross section of the tall needle structures. The tallest features examined on MPEP sample P39 were approximately 1000 nm tall (Fig. 9d). The deepest subsurface porous layer on MP sample P43 830 nm (Fig. 9b) was slightly deeper than the 730 nm deep layer observed on MP sample S25 (Fig. 6b). Previous experiments in the MITE-E on MPEP PCW samples revealed that grains close to $\{100\}$ orientations were less eroded than grains near $\{110\}$ [22]. It is therefore expected that features on the PCW samples with grains near $\{100\}$ would have taller features than the SCW (110) samples.

Sample P37 was MPEP; but after the polishing and before irradiation, an identifying mark approximately $25 \text{ } \mu\text{m}$ wide was scratched into the surface using a diamond-tipped scoring tool (left side of Fig. 10b). Immediately beside the scratched area, the surface has the MPEP surface finish (right side of Fig. 10b). When a material is scratched, the frictional sliding and plowing effects cause plastic deformation and can relocate material from the center to the edges of the trough left by the scoring tool [35]; these effects locally change the microstructure in a similar way as MP. Sample P37 was irradiated to an average fluence of $3.0 \times 10^{22} \text{ He}^+/\text{m}^2$, but the area examined in Fig. 10 is $\sim 1.4 \text{ mm}$ from the center of the damage zone. It can be seen that the morphologies that developed in the scratched region of sample P37 (Fig. 10a) match more closely with the MP sample P42 (Fig. 8b), whereas the morphology that developed on the rest of

the MPEP surface (Fig. 10c) closely matches the MPEP sample P29. Thus it was observed that even local changes to the surface morphology before irradiation cause different morphologies to develop after irradiation.

Fig. 10. Sample P37 was MPEP and then scratched, before irradiation to an average fluence of $3.0 \times 10^{22} \text{ He}^+/\text{m}^2$ with 30 keV He^+ at 1173 K. All images are taken at $\sim 1.4 \text{ mm}$ from the center of the irradiated zone. (a) Within the scratch, a morphology developed similar to the morphology on MP PCW samples. (b) The scratched area appears smeared (left side) immediately adjacent to the smooth MPEP surface finish (right side) before irradiation. (c) The morphology that developed outside the scratch matches the morphology on other MPEP samples to similar conditions.

3.3 Comparison with previous UW-IEC experiments

For previous irradiation experiments using HOMER or HELIOS at the UW-IEC Lab, all tungsten samples were MP before irradiation [20, 36-38]. All samples previously used in the MITE-E were MPEP [7, 21, 22]. It was difficult to make comparisons between the MITE-E work and previous irradiation experiments in HOMER and HELIOS both because of the different irradiation devices and because of the different sample pre-irradiation treatments. The closest comparison with those experiments can be drawn from the MP SCW and PCW samples irradiated in HELIOS and presented in Fig. 4 in Ref. [38]. For both the PCW and SCW samples irradiated in HELIOS, at the lower fluence of $4 \times 10^{21} \text{ }^3\text{He}^+/\text{m}^2$ and 1273 K, the surface was roughened, but the mechanical polish scratches were still visible [38]. At the higher fluence of $5 \times 10^{22} \text{ }^3\text{He}^+/\text{m}^2$ at 1273 K, a coral morphology developed [38]. The coral morphology is a uniformly distributed porous and fibrous structure that was seen on tungsten samples previously irradiated in the HOMER and HELIOS devices [38-41]. The coral morphology is characteristically smaller than the porous structure that developed on sample S18. The coral morphology is uniform across all grain orientations, and grain boundaries are not visible as seen with the grass structure on samples irradiated in the MITE-E [7].

The depths of the subsurface porous layer observed in FIB cross sections for selected samples are compared in Table 2 and listed in order of increasing damage layer thickness. In general, the depth of damage increases with increasing fluence. The samples irradiated in HELIOS all have shallower damage layers than corresponding samples irradiated in the MITE-E. The main exception to the trends is sample S18, the MPEP sample that was irradiated in the MITE-E to the highest fluence but has the shallowest damage layer.

In the HELIOS device, ions bombard the sample from all angles simultaneously, while in the MITE-E, the ions are all normal to the sample surface. The angle of incident ions affects the depth of implantation and is important in determining the morphology that develops. Also, the samples irradiated in the HELIOS device were bombarded with 30 keV $^3\text{He}^+$, which has a

slightly deeper range than 30 keV $^4\text{He}^+$ used in the MITE-E. Although they had the same pre-irradiation surface treatment, the MP samples developed different morphologies and different subsurface damage layers depending on whether they were irradiated in HELIOS or the MITE-E.

Table 2. Comparison of subsurface damage layer depths in mechanically polished (MP) and mechanically polished and electropolished samples (MPEP) before irradiation in either the MITE-E or HELIOS with 30 keV He ions.

Irradiation device	Sample ID	Temperature (K)	Fluence (He/m^2)	Depth of bubbles (nm)	Pre-irradiation treatment
MITE-E	S18	1173	6×10^{22}	90	MPEP
HELIOS	SCW [38]	1273	4×10^{21}	130	MP
HELIOS	PCW [38]	1273	4×10^{21}	140	MP
HELIOS	SCW [38]	1273	5×10^{22}	190	MP
MITE-E	S22	1173	1×10^{22}	250	MP
HELIOS	PCW [38]	1273	5×10^{22}	290	MP
MITE-E	P42	1173	1×10^{22}	350	MP
MITE-E	S25	1173	6×10^{22}	730	MP
MITE-E	P43	1173	6×10^{22}	830	MP
MITE-E	P39	1173	6×10^{22}	1000	MPEP

3.4 Mass loss

Sample masses were measured before and after irradiation and the mass loss was normalized by the irradiated area on the sample (Fig. 11). The mass losses of the SCW MPEP and PCW MPEP sample series used for morphology comparison in Figs. 2 and 8 were reported previously in Refs. [21] and [22], respectively. A second set of PCW MPEP sample mass loss data was published by Zenobia et al. in Ref. [7] and is included because the samples were irradiated under the same conditions in the MITE-E as the other sample discussed in this work. The SCW MPEP samples and the lower two fluence samples of the SCW MP series were weighed with a coating on the back sides of the samples, which resulted in overestimation of the mass they lost because of ion bombardment. This procedure was corrected for later samples, so the other samples in Fig. 11 have smaller uncertainties in the mass loss values.

Fig. 11. Mass losses of single crystal tungsten (SCW) and polycrystalline tungsten (PCW) that had either mechanical polish (MP) or mechanical polish and electropolish (MPEP) before irradiation with 30 keV He⁺ at 1173 K in the MITE-E are compared. ^aRef. [21], ^bRef. [22], ^cRef. [7].

Overall, there is a trend of increasing mass loss with increasing fluence. At the lowest fluence of 3×10^{21} He⁺/m², the morphology of MP SCW sample S23 did not have significant height variations (Fig. 2b). It is expected that sample S23 lost mass primarily because of individual atom sputtering. At this low fluence, a distinction cannot be made between the mass loss of the MP SCW, MPEP SCW, and MPEP PCW samples tested.

At the medium fluence of 1×10^{22} He/m², MP sample S22 shows elevation differences of ~250 nm on the sample surface (Fig. 6a) and likely lost larger sections of material from the lower regions. It appears that the SCW MP sample S22 lost more mass than the SCW MPEP, PCW MP, and PCW MPEP samples; but it cannot be determined conclusively because of the overlapping error bars. It is clear that the surface finish of the PCW samples did not significantly impact the mass losses at this fluence, as all PCW mass loss values are clustered together between ~2 and ~3 g/m².

At a fluence of 1×10^{22} He/m² and higher, mass loss values for all types of tungsten samples are significantly higher than the sputtering mass loss estimate, which was calculated using the ion fluence and a yield of 0.035 tungsten atoms sputtered per incident helium ion [42]. The sputtering estimate is indicated in Fig. 11 by the far right bar at each fluence and a horizontal line at the three highest fluences. At 1×10^{22} He/m², significant helium pore formation was observed on the surface of the samples; and it is thought that samples lost mass when pores opened to release helium gas pressure in a bubble, or that delicate tendrils in the morphology were lost in addition to the single-atom sputtering from the incident ions. Mass loss greater than that from sputtering has been observed by other researchers studying helium implantation in W [43].

At the highest fluence tested of 6×10^{22} He/m², it is clear that the SCW MP sample lost less mass than the other samples discussed in this paper, but slightly more, outside the error bars, mass than the PCW MPEP sample irradiated by Zenobia et al. [7]. This result is interesting

because the morphologies that developed on the SCW MP and PCW MP samples irradiated to the highest fluence are almost identical (Fig. 2d and Fig. 8d). The difference in mass losses for the SCW MP and SCW MPEP samples at the highest fluence was understood by examining the depths of visible bubbles in Table 2, which was 730 nm for the SCW MP sample and 90 for the SCW MPEP sample. It appears that the SCW MP sample retained more tungsten within a subsurface semi-porous layer and had a mass loss of $\sim 7 \text{ g/m}^2$; whereas the SCW MPEP sample lost more material from the original surface, resulting in a $\sim 15 \text{ g/m}^2$ mass loss and a shallower bubble layer.

As was the case at $1 \times 10^{22} \text{ He/m}^2$, at $6 \times 10^{22} \text{ He/m}^2$, the PCW MP and the PCW MPEP samples lost essentially the same amount of mass. However, at that high fluence, these PCW samples lost significantly more mass than the sample irradiated by Zenobia et al. in Ref. [7] to the same conditions. The difference in mass loss between the samples discussed here and those of Zenobia et al. [7] may be attributed to the statistical nature of the experiment and slight differences in sample handling procedures between the experiments.

Mass loss of tungsten can be detrimental to any fusion reactor by either negatively impacting the plasma or destroying the plasma-facing component. Although only slight differences in mass loss were observed between the different orientations and different pre-irradiation surface treatments, an optimal surface finish could result in a significant reduction in mass loss for the plasma-facing components when multiplied over the 210 m^2 area of the ITER divertor [44] or the $\sim 1450 \text{ m}^2$ area of the reference HAPL chamber [3]. The mass loss and morphology formation are sensitive to the incoming ion energy, so to determine the best surface finish of tungsten for a particular fusion reactor would require exposure to prototypical reactor plasma conditions.

4. Conclusions

The SCW sample that was MP before irradiation in the MITE-E with 30 keV He^+ ions at the highest fluence of $6.0 \times 10^{22} \text{ He}^+/\text{m}^2$ at 1173 K developed a grass morphology 730 nm deep. The SCW sample that was MPEP developed a shallow, 90 nm deep, large pore structure at the same irradiation conditions. The difference in morphology corresponded to a difference in mass loss: the MPEP SCW sample lost more than twice as much mass as the MP SCW sample.

The morphology progression on the MP PCW samples was similar to that on the MP SCW samples as the fluence increased. At medium fluence, both samples developed patches of higher and lower morphologies; and at the highest fluence of $6.0 \times 10^{22} \text{ He}^+/\text{m}^2$ at 1173 K, both developed a grass structure. However, for the MP PCW sample, scattered over the grass structure were remnants of porous needle structures. The needle structures were mostly fragmented but were up to 800 nm long. The MP PCW and MPEP PCW samples had different morphologies at intermediate fluences; but at the highest fluence, they had similar morphologies except for the porous needles on the MP PCW sample. There was not a noticeable difference in mass loss between the PCW samples with different pre-irradiation surface finishes.

Neither the MP SCW nor the MP PCW samples irradiated in the MITE-E resembled the respective MP SCW or MP PCW samples irradiated previously in HELIOS to similar fluences [38]. This indicates that for these samples, the method of ion irradiation had more influence on the morphology than the pre-irradiation surface finish.

Although similar regions could be found on the MP PCW and MPEP PCW samples irradiated to the highest fluence, the morphology progression with increasing fluence was

different for each pre-irradiation surface type. In comparing the MP and MPEP SCW samples, it would be impossible to determine by observation alone that they were irradiated to the same conditions. These results show that sample surface preparation before irradiation can have a dramatic effect on the morphology that develops after irradiation. Although neither surface finish prevented significant mass loss, there is room for improvement in optimizing the surface for use in fusion reactors. For experiments studying the effects of reactor-like conditions on tungsten, surface preparation methods should be carefully documented and reported so that the best comparisons can be made among different experiments.

Acknowledgements

This research is sponsored in part by the Office of Fusion Energy Sciences, U.S. Department of Energy (U.S. DOE), under contract DE-AC05-00OR22725 with UT-Battelle, LLC and U.S. DOE Office of Science Graduate Fellowship Program, made possible in part by the American Recovery and Reinvestment Act of 2009, administered by ORISE-ORAU under contract no. DE-AC05-06OR23100. Additional financial support was provided by the University of Wisconsin–Madison, the Grainger Foundation, and the Greatbatch Foundation. The authors would like to thank Oak Ridge National Laboratory for providing the samples used in these experiments and the UW-IEC team for their assistance. Chad Parish, Yutai Katoh, and Deborah Counce are thanked for their assistance with the manuscript.

References

- [1] V. Barabash, The ITER International Team, A. Peacock, S. Fabritsiev, G. Kalinin, S.J. Zinkle, A. Rowcliffe, J.-W. Rensman, A.A. Tavassoli, P. Marmy, P.J. Karditsas, F. Gillemot, M. Akiba, J. Nucl. Mater., 367-370 (2007) 21-32.
- [2] ITER Newslines, Council Takes Important Technical and Managerial Decisions, (2013), <http://www.iter.org/newslines/291/1777>, 3/3/2014
- [3] J.D. Sethian, A.R. Raffray, J. Latkowski, J.P. Blanchard, L. Snead, T.J. Renk, S. Sharafat, J. Nucl. Mater., 347 (2005) 161-177.
- [4] Y. Ueda, M. Fukumoto, J. Yoshida, Y. Ohtsuka, R. Akiyoshi, H. Iwakiri, N. Yoshida, J. Nucl. Mater., 386-388 (2009) 725-728.
- [5] M.J. Baldwin, R.P. Doerner, Nucl. Fusion, 48 (2008) 1-5.
- [6] A.I. Ryazanov, V.S. Koidan, B.I. Khripunov, S.T. Latushkin, V.B. Petrov, L.S. Danelyan, E.V. Semenov, V.N. Unezhev, Fusion Sci. Technol., 61 (2012) 107-117.
- [7] S.J. Zenobia, L.M. Garrison, G.L. Kulcinski, J. Nucl. Mater., 425 (2012) 83-92.
- [8] J.-Z. Jiang, Y.-W. Zhao, Y.-G. Wang, J.-B. Luo, Wear, 265 (2008) 992-998.
- [9] L.E. Samuels, Chapter 8 Polishing with Abrasives: Surface Damage, Metallographic Polishing by Mechanical Methods, fourth ed., ASM International, Materials Park, OH, 2003.
- [10] L.E. Samuels, Chapter 5 Machining with Abrasives: Surface Damage, Metallographic Polishing by Mechanical Methods, fourth ed., ASM International, Materials Park, OH, 2003.
- [11] S.L. Rice, H. Nowotny, S.F. Wayne, Wear, 74 (1981) 131-142.
- [12] L.E. Samuels, Metallographic Polishing by Mechanical Methods, fourth ed., ASM International, Materials Park, OH, 2003.
- [13] D.A. Rigney, J.P. Hirth, Wear, 53 (1979) 345-370.

- [14] L.E. Samuels, Chapter 3 Machining with Abrasives: Principles, Metallographic Polishing by Mechanical Methods, fourth ed., ASM International, Materials Park, OH, 2003.
- [15] E. Lassner, W.-D. Schubert, Tungsten, Kluwer Academic/Plenum Publishers, New York, 1999.
- [16] L.E. Samuels, Chapter 7 Polishing with Abrasives: Principles, Metallographic Polishing by Mechanical Methods, fourth ed., ASM International, Materials Park, OH, 2003.
- [17] D.A. Rigney, W.A. Glaeser, Wear, 46 (1978) 241-250.
- [18] L.E. Samuels, Chapter 11 Modifications Required to Manual Preparation Systems, Metallographic Polishing by Mechanical Methods, fourth ed., ASM International, Materials Park, OH, 2003.
- [19] U. Samm, Fusion Sci. Technol., 61 (2012) 193-198.
- [20] R.F. Radel, G.L. Kulcinski, J. Nucl. Mater., 367-370 (2007) 434-439.
- [21] L.M. Garrison, G.L. Kulcinski, Fusion Sci. Technol., 64 (2013) 216-220.
- [22] L.M. Garrison, G.L. Kulcinski, Phys. Scripta, T159 (2014) 014020.
- [23] L.E. Samuels, Chapter 9 Brittle Materials: Principles, Metallographic Polishing by Mechanical Methods, fourth ed., ASM International, Materials Park, OH, 2003.
- [24] U. Valbusa, C. Boragno, F. Buatier de Mongeot, J. Phys.: Condens. Matter, 14 (2002) 8153-8175.
- [25] G. Antczak, G. Ehrlich, Surf. Sci. Rep., 62 (2007) 39-61.
- [26] D. Nishijima, M.Y. Ye, N. Ohno, S. Takamura, J. Nucl. Mater., 329-333 (2004) 1029-1033.
- [27] K. Hermann, Surface Explorer Version 2, Based on Balsac©, Fritz Haber Institute, <http://surfexp.fhi-berlin.mpg.de/>
- [28] J.M. Colino, M.A. Arranz, Applied Surface Science, 257 (2011) 4432-4438.
- [29] A. Toma, D. Chiappe, B.S. Batic, M. Godec, M. Jenko, F.B. De Mongeot, Physical Review B (Condensed Matter and Materials Physics), 78 (2008) 153404-153406.
- [30] G.P. Thomson, Structure and Properties of Solid Surfaces, The University of Chicago Press, Chicago, IL, 1953.
- [31] G. Ehrlich, Surf. Sci., 246 (1990) 1-12.
- [32] M. Rösenberg, H. Viehhaus, Surf. Sci., 172 (1986) 615-640.
- [33] T.J. C., J.M. Blakely, Surf. Sci., 13 (1969) 313-336.
- [34] M. Hou, C.J. Ortiz, C.S. Becquart, C. Domain, U. Sarkar, A. Debacker, J. Nucl. Mater., 403 (2010) 89-100.
- [35] Z. Liu, J. Sun, W. Shen, Tribol. Int., 35 (2002) 511-522.
- [36] B.B. Cipiti, G.L. Kulcinski, J. Nucl. Mater., 347 (2005) 298-306.
- [37] R.F. Radel, G.L. Kulcinski, Fusion Sci. Technol., 52 (2007) 544-548.
- [38] S.J. Zenobia, G.L. Kulcinski, Fusion Sci. Technol., 56 (2009) 352-360.
- [39] S.J. Zenobia, R.F. Radel, B.B. Cipiti, G.L. Kulcinski, J. Nucl. Mater., 389 (2009) 213-220.
- [40] S.J. Zenobia, G.L. Kulcinski, Phys. Scripta, T138 (2009) 014049.
- [41] S.J. Zenobia, L.M. Garrison, G.L. Kulcinski, Fusion Sci. Technol., 60 (2011) 344-348.
- [42] R. Behrisch, W. Eckstein, Sputtering by Particle Bombardment, Springer, Berlin, 2007.
- [43] H. Maier, H. Greuner, M. Balden, B. Böswirth, S. Elgeti, U.v. Toussaint, C. Linsmeier, Phys. Scripta, T159 (2014) 014019.
- [44] M. Merola, F. Escourbiac, R. Raffray, P. Chappuis, T. Hirai, A. Martin, Fusion Eng. Des., 89 (2014) 890-895.